

Relativistic corrections to transition frequencies of Ag I, Dy I, Ho I, Yb II, Yb III, Au I and Hg II and search for variation of the fine structure constant

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Dependence of transition frequencies on the fine structure constant $\alpha = e^2/\hbar c$ is calculated for several many-electron systems which are used or planned to be used in the laboratory search for the time variation of the fine structure constant. In systems with a large number of electrons in open shells (from 11 to 15) the relative effects of the variation may be strongly enhanced. For the transitions which were considered before the results are in good agreement with previous calculations.

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I. INTRODUCTION

Theories unifying gravity with other interactions as well as many cosmological models allow for space-time variation of fundamental constants. Experimental search for the manifestation of this variation spans the whole lifetime of the Universe from Big Bang nucleosynthesis to the present-day very precise atomic clock experiments (see, e.g. reviews [1, 2]). An evidence that the fine-structure constant might be smaller about ten billion years ago was found in the analysis of quasar absorption spectra [3, 4, 5, 6, 7, 8]. This finding together with progress in developing of very precise atomic frequency standards motivated many laboratory searches for the present-day time variation of the fundamental constants (see, e.g. [9]). In particular, strong limit on the rate of the time variation of the fine structure constant α ($\alpha = e^2/\hbar c$) were found by comparing frequencies of different atomic transitions over few years [9].

Apart from the microwave atomic clocks and optical frequency standards, a number of atomic transitions in which the change of frequency due to change of α is strongly enhanced has been suggested in Refs. [10, 11, 12].

Interpretation and planning of the measurements of the α variation require atomic calculations to relate the change of atomic frequencies to the change of the fine structure constant. A number of such calculations for atomic optical transitions have been performed in our early works [10, 11, 12, 13, 14, 15]. Independent calculations for some optical transitions have been recently reported in Ref. [16].

From the computational point of view the most important parameter of an atom which determines the choice of the computational method as well as the accuracy which can be achieved in the calculations is the number of electrons in open shells. The larger the number the more difficult are the calculations. Many optical frequency standards are based on atoms or ions with just one or two valence electrons [9]. Calculations for such systems are accurate and reliable [10, 13, 15]. However, many atomic systems which are used or planned to be used in laboratory search for variation of the fine

structure constant have more than ten electrons in open shells. For example, strong limits on the variation of α in time [17] and variation of α due to change of the gravitation potential [18] were obtained with the use of dysprosium atom which has twelve external electrons (see also [10, 14, 19]). There are plans to use holmium (13 electrons) for similar measurements [20]. There are ongoing measurements or plans for measurements for Ag I [21], Yb II, [22, 23], Yb III [24] and Hg II [25] (see also a review [9] and references therein). These systems involve states with excitations from d or f subshells and therefore must be treated as many-valence-electrons systems.

Calculations for many-valence-electron atoms are difficult due to the fast growth of the matrix size of the configuration interaction (CI) eigenvalue problem with the increase of the single-electron basis. In our recent paper on Fe I [26] we used a version of the CI method which is similar to multi-configuration CI method (see, e.g. Ref. [27]) and which allows to obtain reasonably accurate result with a very short basis. In present paper we use this method for many-electron systems which are of the interest for laboratory search of the variation of the fine structure constant. The aim of the calculation is to check our early results as well as to calculate relativistic energy shifts for atomic transitions which have never been considered before.

II. METHOD

Detailed discussion of the method can be found in our early works [10, 26]. Here we repeat its major points.

It is convenient to present the dependence of atomic frequencies on the fine-structure constant α in the vicinity of its physical value α_0 in the form

$$\omega(x) = \omega_0 + qx, \quad (1)$$

where ω_0 is the present laboratory value of the frequency and $x = (\alpha/\alpha_0)^2 - 1$, q is the coefficient which is to be found from atomic calculations. Note that

$$q = \left. \frac{d\omega}{dx} \right|_{x=0}. \quad (2)$$

To calculate this derivative numerically we use

$$q \approx \frac{\omega(+\delta) - \omega(-\delta)}{2\delta}. \quad (3)$$

In the present calculations we use $\delta = 0.05$, which leads to

$$q \approx 10 (\omega(+0.05) - \omega(-0.05)). \quad (4)$$

In a single-electron approximation relativistic energy shift can be estimated using the formula [10]

$$\Delta_a = \frac{E_a}{\nu_a} (Z\alpha)^2 \left[\frac{1}{j_a + 1/2} - C(Z, j_a, l_a) \right], \quad (5)$$

where a is the index for a single-electron state, E_a is its energy, ν_a is its effective principal quantum number ($\nu_a = 1/\sqrt{-2E_a}$), j_a and l_a are total and angular momenta of the state a . $C(Z, j_a, l_a)$ is a parameter which is introduced to simulate the effect of Hartree-Fock exchange interaction and other many-body effects. For a transition between many-electron states which can be approximated as a single-electron transition from state a in lower level to state b in upper level one has

$$q \approx \Delta_b - \Delta_a. \quad (6)$$

The formulas (5),(6) are too inaccurate for practical use in the interpretation of the measurements. However, they are very useful for predicting what one can expect to find in different atomic transitions and for explaining the values and sign of the relativistic corrections. We will use it for the discussion of our results.

For accurate numerical calculations of the coefficients q using (4), α must be varied in the computer code. Therefore, it is convenient to use a form of the single electron wave function in which the dependence on α is explicitly shown (we use atomic units in which $e = \hbar = 1, \alpha = 1/c$)

$$\psi(r)_{njlm} = \frac{1}{r} \begin{pmatrix} f_v(r)\Omega(\mathbf{n})_{jlm} \\ i\alpha g_v(r)\tilde{\Omega}(\mathbf{n})_{jlm} \end{pmatrix}, \quad (7)$$

where n is the principal quantum number and an index v replaces the three-number set n, j, l . This leads to a form of radial equation for single-electron orbitals which also explicitly depends on α :

$$\begin{aligned} \frac{df_v}{dr} + \frac{\kappa_v}{r} f_v(r) - \left[2 + \alpha^2(\epsilon_v - \hat{V}_{HF}) \right] g_v(r) &= 0, \\ \frac{dg_v}{dr} - \frac{\kappa_v}{r} f_v(r) + (\epsilon_v - \hat{V}_{HF}) f_v(r) &= 0, \end{aligned} \quad (8)$$

here $\kappa = (-1)^{l+j+1/2}(j+1/2)$, and \hat{V}_{HF} is the Hartree-Fock potential. Equation (8) with $\alpha = \alpha_0\sqrt{\delta+1}$ and different Hartree-Fock potential \hat{V}_{HF} for different configurations is used to construct single-electron orbitals.

Table I lists configurations considered in present work. For Ag I, Au I and Hg II we use only ground-state configuration and configurations, involving excitation from the upper core d -state. The latter corresponds to the states

TABLE I: Configurations and effective core polarizabilities (α_p , a.u.) used in the calculations.

Atom	Z	N_v^a	Set	Parity	Configuration	α_p
Ag I	47	11	1	Even	$4d^{10}5s$	0.4
			2	Even	$4d^95s^2$	0.414
Dy I	66	12	1	Even	$4f^{10}6s^2$	0.4
			2	Even	$4f^{10}5d6s$	0.397
			3	Even	$4f^96s^26p$	0.4039
			4	Even	$4f^95d6s6p$	0.389
			5	Odd	$4f^95d^26s$	0.3895
			6	Odd	$4f^95d6s^2$	0.4
			7	Odd	$4f^{10}6s6p$	0.393
			1	Odd	$4f^{11}6s^2$	0.4
			2	Odd	$4f^{10}6s^26p$	0.401
			3	Odd	$4f^{11}5d6s$	0.401
Ho I	67	13	4	Odd	$4f^{10}5d6s6p$	0.39
			5	Odd	$4f^{11}6p^2$	0.39
			6	Even	$4f^{10}5d6s^2$	0.3927
			7	Even	$4f^{11}6s6p$	0.3962
			8	Even	$4f^{10}5d^26s$	0.39
			9	Even	$4f^{10}5d6p^2$	0.4
			10	Even	$4f^{10}6s6p^2$	0.4
			1	Even	$4f^{14}6s$	0.4
			2	Even	$4f^{13}6s^2$	0.399
			3	Even	$4f^{13}5d6s$	0.3911
Yb II	70	15	4	Even	$4f^{13}5d^2$	0.39
			1	Even	$4f^{14}$	0.4
			2	Even	$4f^{13}5d$	0.3914
Yb III	70	14	3	Even	$4f^{13}6s$	0.3977
			1	Even	$5d^{10}6s$	0.4
			2	Even	$5d^96s^2$	0.417
Au I	79	11	1	Even	$5d^{10}6s$	0.4
			2	Even	$5d^96s^2$	0.417
Hg II	80	11	1	Even	$5d^{10}6s$	0.4
			2	Even	$5d^96s^2$	0.426

^a N_v is the number of valence electrons.

which are to be used in the measurements. We add more configurations for Yb II and Yb III and even more for Dy I and Ho I. In the latter atoms the states of interest are highly excited ones for which configuration mixing is strong and should be taken into account more accurately.

The self-consistent Hartree-Fock procedure is done for every configuration listed in Table I separately. Then valence states found in the Hartree-Fock calculations are used as basis states for the CI calculations. It is important for the CI method that the atomic core remains the same for all configurations. We use the core which corresponds to the ground state configuration. Change in the core due to change of the valence state is small and can be neglected. This is because core states are not sensitive to the potential from the electrons which are on large distances (like $6s$, $6p$ and $5d$ electrons). The $4f$ electrons are on smaller distances and have larger effect on atomic core. However, in all the cases (see Table I) only one among about ten $4f$ electrons change its state. Therefore their effect on atomic core is also small. More detailed discussion of the effect of valence electrons on atomic core can be found in Refs. [28, 29].

The effective Hamiltonian for N_v valence electrons has the form

$$\hat{H}^{\text{eff}} = \sum_{i=1}^{N_v} \hat{h}_{1i} + \sum_{i<j}^{N_v} e^2/r_{ij}, \quad (9)$$

$\hat{h}_1(r_i)$ is the one-electron part of the Hamiltonian

$$\hat{h}_1 = c\alpha \cdot \mathbf{p} + (\beta - 1)mc^2 - \frac{Ze^2}{r} + V_{\text{core}} + \delta V. \quad (10)$$

Here α and β are Dirac matrixes, V_{core} is Hartree-Fock potential due to core electrons and δV is the term which simulates the effect of the correlations between core and valence electrons. It is often called *polarization potential* and has the form

$$\delta V = -\frac{\alpha_p}{2(r^4 + a^4)}. \quad (11)$$

Here α_p is polarization of the core and a is a cut-off parameter (we use $a = a_B$).

The form of the δV is chosen to coincide with the standard polarization potential on large distances ($-\alpha_p/2r^4$). However we use it on distances where valence electrons are localized. This distances are not large, especially for the $4f$ electrons. Therefore we consider δV as only rough approximation to real correlation interaction between core and valence electrons and treat α_p as fitting parameters. The values of α_p for each configuration of interest are presented in Table I. They are chosen to fit the experimental position of the configurations relative to each other. For all configurations of the same atom the values of α_p are very close. This is not a surprise since the core is nearly the same for all configurations of interest. One can probably say that small difference in α_p for different configurations simulates the effect of incompleteness of the basis and other imperfections in the calculations.

Tables II and III present comparison between experimental and theoretical energies and g -factors for Dy I and Ho I atoms. The g -factors are useful for the identification of the states and for control of configuration mixing [30]. For dysprosium atom both the energies and g -factors are reproduced quite accurately. This includes the states with the energies of 19797.96 cm^{-1} which are used in the measurements [17, 18].

For holmium the g -factors are not known for the most of the states. This makes it more difficult to identify the states and to judge about the accuracy of the calculations of the relativistic energy shifts. If the measurements for holmium are to go ahead it would be good to measure the g -factors as well, at least for the states of most interest. At the moment we can only rely on the energies. Although the energies are reproduced in the calculations quite accurately the coefficients q in (1) are very sensitive to the configuration mixing which in turn is sensitive to the energy intervals between close levels of the same parity and total angular momentum. Therefore, having good

TABLE II: Energy levels (cm^{-1}) and g -factors of some low states of Dy I

Conf.	J	Experiment ^a		Calculations	
		Energy	g	Energy	g
$4f^{10}6s^2$	8	0.00	1.242	0	1.2428
	7	4134.23	1.173	4409	1.1747
	6	7050.61	1.072	7600	1.0723
	5	9211.58	0.911	9983	0.9080
	4	10925.25	0.618	11840	0.6163
$4f^{10}5d6s$	9	17514.50	1.316	17703	1.3145
	8	18903.21	1.22	19556	1.2754
	7	21074.20	1.24	21881	1.1983
	8	17613.36	1.33	17871	1.3300
	7	18937.78	1.28	19633	1.3012
	6	21159.79	1.24	22042	1.2116
	7	18094.52	1.38	18308	1.3835
	6			20090	1.3078
	5			22478	1.2198
	10	18462.65	1.282	18461	1.2883
$4f^{10}6s^2$	9	19240.82	1.217	19592	1.2277
	8	20193.60	1.16	20893	1.1700
	8	19019.15	1.14	21377	1.1113
$4f^{10}5d6s$	11	19348.72	1.27	19295	1.2675
	10	19797.96 ^b	1.21	20077	1.2089
$4f^96s^26p$	9	20209.00	1.14	20847	1.1261
	7	20614.32	1.32	19835	1.3372
$4f^{10}5d6s$	8	20789.85	1.32	19832	1.2997
	8	21603.04	1.26	23205	1.2514
$4f^95d6s6p$	7	21778.43	1.26	23232	1.2419
	9	22045.79	1.22	23429	1.2677
	10	22487.14	1.197	24132	1.2162
$4f^95d6s^2$	8	23031.46	1.37	23132	1.3730
$4f^{10}6s6p$	10	12892.76	1.29	12920	1.2933
$4f^{10}6s6p$	10	17513.33	1.30	17582	1.2944
$4f^95d^26s$	10	19797.96 ^b	1.367	19693	1.3677
$4f^95d^26$	10	21788.93	1.34	22312	1.3340

^aNIST, Ref. [31]

^bStates used in the measurements [17, 18]

accuracy for absolute values of energies is not enough for reliable results for the coefficients q . It is very important that the relative positions of the states around the states of interest are reproduced accurately in the calculations.

III. RESULTS AND DISCUSSION

a. Holmium Holmium atom has been suggested for the search of the variation of the fine structure constant by Mark Saffman [20]. From the computational point of view it represents the most difficult case. It has thirteen electrons in open shells, very dense spectrum, strong configuration mixing and multiple level crossing in the vicinity of the physical value of α when energies are considered as functions of α^2 . All these factors contribute to the instability of the results. Therefore, it is instructive to start from simple estimations based on a single-electron approximation. Table IV shows approximate values of

TABLE III: Energy levels (cm^{-1}) and g -factors of some low states of Ho I

Conf.	Parity	J	Expt. ^a Energy	Calculations Energy	g
$4f^{11}6s^2$	Odd	15/2	0.00	0	1.20
		13/2	5419.70	5770	1.11
$4f^{10}6s^26p$	Odd	15/2	18572.28	18343	1.28
$4f^{11}5d6s$	Odd	13/2	18867.40	18684	1.37
		15/2	19276.94	19295	1.32
$4f^{10}5d6s6p$	Odd	15/2	24112.04	23908	1.35
$4f^{10}5d6s^2$	Even	15/2	8427.11	8395	1.28
		13/2	9147.08	9341	1.33
$4f^{10}5d6s^2$	Even	15/2	12339.04	12903	1.23
		13/2	12344.55	12953	1.23
		11/2	13082.93	13799	1.25
		13/2	15081.12	16459	1.17
$4f^{11}6s6p$	Even	11/2	16937.43	16817	1.13
		15/2	15855.28	15913	1.28
$4f^{10}5d^26s$	Even	13/2	17059.35	17135	1.20
		15/2	20167.17	20138	1.41

^aNIST, Ref. [31]

TABLE IV: Approximate values of the q -coefficients for different configurations of Ho I ($\times 10^3 \text{ cm}^{-1}$).

Configuration	Parity	Transition ^a	q
$4f^{11}6s^2$	Odd	(ground state)	5(5)
$4f^{10}5d6s^2$	Even	$4f \rightarrow 5d$	-35(15)
$4f^{11}6s6p$	Even	$6s \rightarrow 6p$	4(4)
$4f^{10}6s^26p$	Odd	$4f \rightarrow 6p$	-45(15)
$4f^{11}5d6s$	Odd	$6s \rightarrow 5d$	7(4)

^asingle-electron transition from the ground state.

q -coefficients for different configurations of holmium obtained with the use of formula (6) but with energy shifts of the individual single-electron states ($\Delta_{a,b}$) taken from the Hartree-Fock calculations rather than from formula (5). Note that we present the energies and q -coefficients relative to the ground state. Therefore, relativistic energy shift (q) for the ground state is zero by definition. The q -coefficients for other states of the $4f^{11}6s^2$ configuration are determined by the fine structure of the $4f$ orbital. Large error bars are due to the fact that relativistic energy shifts depend on the values of the total momentum j of the single-electron states involved in the transition (see formula (5)). For example, the $6s \rightarrow 6p$ transition can be the $6s \rightarrow 6p_{1/2}$ or the $6s \rightarrow 6p_{3/2}$ transition, etc.

As can be seen from the data in Table IV the values of q are very different for different configurations. For example, $q \approx -35000 \text{ cm}^{-1}$ for the $4f^{10}5d6s^2$ configuration and $q \approx 4000 \text{ cm}^{-1}$ for the $4f^{11}6s6p$ configuration. But these two configurations have the same parity and can have the states of the same total angular momentum J . Therefore, if these configurations are strongly mixed

the resulting values of q will be linear combinations of $q \approx -35000 \text{ cm}^{-1}$ and $q \approx 4000 \text{ cm}^{-1}$, i.e. they may take any value depending on which configuration dominates in the state. The same is true for odd states which constitute a mixture of the negatively shifted $4f^{10}6s^26p$ configuration with the positively shifted $4f^{11}5d6s$ or $4f^{11}6s^2$ configurations. The analysis of the holmium spectrum shows that there are many states of the same parity and total angular momentum J which are separated by only small energy intervals and in which different configurations dominate. These states are strongly mixed which leads to instability of the calculations of the q -coefficients. The only way to obtain reliable results is to make sure that the relative position of the states in the vicinity of state of interest as well as the energy intervals between these states are reproduced accurately in the calculations. This can be achieved by appropriate choice of the α_p parameters for different configurations (see Table I).

In Table V we present the results of the calculations for two pairs of almost degenerate states of holmium. The relative change of frequency between degenerate levels due to change of α is strongly enhanced by small energy interval. The enhancement factor K (defined by $\delta\omega/\omega = K\delta\alpha/\alpha$ where ω is the transition frequency) is given by [10]

$$K = 2\Delta q / \Delta E. \quad (12)$$

This enhancement factor is about 3×10^5 for both pairs of holmium states presented in Table V. To avoid misunderstanding we should note that the enhancement of the relative effect here is due to the small ΔE ; there is no any enhancement of the absolute values of the frequency shifts. The values of q in holmium are typical for heavy atoms.

b. Dysprosium Dysprosium atom is used for the search of time-variation of the fine structure constant at Berkeley [17, 18, 19]. It has two almost degenerate levels of opposite parity at $E = 19797.96 \text{ cm}^{-1}$ for which the enhancement factor (12) is about 10^8 [14]. Limits on the rate of changing of α in time obtained from monitoring the frequency of the transition between these two levels over long period of time is on the same level of precision as for the most advances atomic optical clock experiments ($\sim 10^{-15}/\text{yr}$) [17]. The interpretation of the measurements are based on our early calculations [14]. The aim of present calculations is to check our previous result with a significantly different method.

From the computational point of view dysprosium atom is an easier case than holmium in two ways. First, it has one less valence electron and its spectrum is much less dense. The energy separation between mixing states is larger than 1000 cm^{-1} which is much easier to reproduce in the calculations than few hundred cm^{-1} as in the case of holmium. Second, experimental values for g -factors are available for dysprosium. The g -factors are almost as sensitive to configuration mixing as the q -coefficients [30] providing an important test of the accuracy of the calcu-

TABLE V: Experimental and theoretical energies and calculated relativistic energy shifts (q -coefficients, cm^{-1}) for some transitions of Ag I, Dy I, Ho I, Yb II, Yb III, Au I and Hg II.

Atom	Ground state		Excited state		Energy [cm^{-1}]		q -coefficients, [cm^{-1}]	
	Conf.	J	Conf.	J	Expt. ^a	this work	this work	other
Ag I	$4d^{10}5s$	$1/2$	$4d^9 5s^2$	$5/2$	30242.26	30188	-11300	
			$4d^9 5s^2$	$3/2$	34714.16	35114	-6500	
Dy I	$4f^{10}6s^2$	8	$4f^{10}5d6s$	10	19797.96	20077	7952	6008 ^b
			$4f^9 5d^2 6s$	10	19797.96	19693	-25216	-23708 ^b
Ho I	$4f^{11}6s^2$	$15/2$	$4f^{10}5d6s^2$	$11/2$	20493.40	21763	-28200	
			$4f^{11}5d6s$	$13/2$	20493.77	20872	7300	
			$4f^{11}6s6p$	$13/2$	22157.86	22599	3000	
			$4f^{11}5d6s$	$9/2$	22157.88	22631	8000	
Yb II	$4f^{14}6s$	$1/2$	$4f^{13}6s^2$	$7/2$	21418.75	20060	-63752	-56737 ^b
			$4f^{13}6s^2$	$5/2$	31568.08	31303	-53400	
			$4f^{13}5d6s$	$5/2$	26759.02	26781	-46863	
			$4f^{13}6s6p$	$7/2$	47921.31	47927	-60432	
Yb III	$4f^{14}$	0	$4f^{13}5d$	0	45276.85	46505	-32800	-27800 ^b
Au I	$5d^{10}6s$	$1/2$	$5d^9 6s^2$	$5/2$	9161.3	9186	-38550	
			$5d^9 6s^2$	$3/2$	21435.3	22224	-26760	
Hg II	$5d^{10}6s$	$1/2$	$5d^9 6s^2$	$5/2$	35514.624	35066	-52200	-56670 ^c
			$5d^9 6s^2$	$3/2$	50555.567	50886	-37700	-44000 ^c

^aNIST, Ref. [31]

^bDzuba *et al.*, Ref. [14]

^cDzuba *et al.*, Ref. [10]

lation. As can be seen from Table II both energies and q -factors are reproduced in the calculations with good accuracy.

The results for the q -coefficients are compared in Table V with previous calculations. The largest relative difference is for the smaller coefficient and is about 30%. However, the difference for Δq which is important for the interpretation of the measurements is only 12%.

c. Ag I, Yb II, Yb III, Au I and Hg II The Ag I, Yb II, Yb III and Hg II atoms are also used or considered for the use in the laboratory search for the variation of the fine structure constant (see [9] and references therein). We have included Au I because it has electron structure similar to Ag I and Hg II. However, we are unaware about any plans to use Au in the measurements.

All these systems utilize the use of a transition from the ground state to a low lying state which involves an excitation from the core. Both states have no significant admixture of other configurations, relatively easy to calculate and produce stable results.

The results for the q -coefficients for the transitions of interest are presented in Table V. Here we also have good

agreement with previous calculations for the cases when the data are available.

IV. CONCLUSION

Calculations of the relativistic energy shifts are presented for many transitions in many-valence-electrons systems which are used or planned to be used in the laboratory search for variation of the fine structure constant. Good agreement with previous calculations confirms the analysis based on old results and provides an estimate of the accuracy of the calculations. Many atomic transitions are added which were never considered before.

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